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Heavy Metal Exposure from Foods

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The Food and Drug Administration has a continuing program of monitoring foods for their content of lead, cadmium, mercury, zinc, arsenic, and selenium to determine trends of increasing or decreasing levels. The monitoring protocol is that of the Total Diet Study, in which "market baskets" of typical foods and beverages consumed by 15- to 20-year-old American males are collected in various geographical locations at regular intervals during the year, divided into food classes, composited, and analyzed. Cadmium has the most widespread distribution of the six heavy metals and mercury the most limited. The analytical values for lead may be underestimated because of limitations of the methodology; these do not apply to the other five elements. A tabulation by year shows that the levels of these elements in foods do not vary significantly from one year to the next. Average intakes of lead, cadmium, and mercury are below the WHO/FAO tolerable intakes for adults; such tolerable intakes have not been established for arsenic and selenium. Increases in concentrations of these elements in foods would be considered undesirable, however.

Determination of the heavy metal content of foods has been carried out by various laboratories of the Food and Drug Administration for a number of years. The data discussed were obtained from the Total Diet Survey program, or as it is better known, the Market Basket Survey. The program was originally designed to monitor the levels of various pesticides in foods and has been ongoing since 1965. In the last few years it has been expanded to include analyses for six heavy metals: lead, cadmium, mercury, zinc, arsenic, and selenium.† Lead, zinc, and selenium have been determined only since 1972, whereas data from earlier years are also available for the other three metals. A major purpose of this program has been to determine the levels of heavy metals in foods, or more specifically, to determine trends of increasing or decreasing levels of metals in various foods or food classes.

The foods included in the Total Diet Study were determined by using 1965 diet survey data of the United States Department of Agriculture which indicate the food consumed by 15- to 20-yr-old American males (*1*). Food consumption of this group was selected for study because the members of the group consume the largest number of calories of any age-sex group in the population, and the largest consumers of food generally have the highest exposure to food contaminants. However, it is recognized that there are possible exceptions to this generality if a food contaminant were present in only a few types of foods.

By using this data base it has been found that over a 14-day period the average diet of young teenage males includes 117 food items. This group of 117 foods is called the Market Basket. These Market Baskets are purchased from retail stores 30 times a year from representative U.S. cities, representing the entire nation within the broadly defined geographic regions, namely, Southeast, Northeast, Central and Western. Thus, the trace metals values to be presented are an average of 30 Market Baskets that are representative of the total food intake in the U.S. in a particular year. Foods which are normally prepared in any manner before consumption are sent to a dietetic kitchen for preparation; for example, meats to be cooked, vegetables to be boiled if they are consumed that way, etc., as indicated by the diet survey data. The quantities of the foods in the Market Basket are based on the USDA survey data.

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†In this context, the term "heavy metals" is used in a colloquial sense and includes the toxic nonmetals arsenic and selenium.

The foods are not analyzed individually but as part of a composite food class, as shown in Table 1. The actual composition of the garden fruits class is shown in Table 2. In all cases portions of the individual samples are retained. In actual practice, more than enough of each food item is collected for the requirements of the 14-day composite. If the level of a contaminant in a composite is unusually high the individual components of the composite will be analyzed to determine the source. Although it may be desirable to analyze all the foods individually, the costs would be very greatly increased.

Table 1. Daily food intake.

	Food class	Food consumption[a] Avg g/day consumed	% of total diet
I	Dairy products	756	25.9
II	Meats, fish, and poultry	290	9.9
III	Grain and cereal products	369	12.6
IV	Potatoes	204	7.0
V	Leafy vegetables	59	2.0
VI	Legume vegetables	74	2.5
VII	Root vegetables	34	1.2
VIII	Garden fruits	88	3.0
IX	Fruits	217	7.4
X	Oils, fats, and shortenings	52	1.8
XI	Sugar and adjuncts	82	2.8
XII	Beverages (including water)	697	23.9
	Total	2922	

[a]Based on FY 1973 Total Diet Survey data of the Food and Drug Administration.

Table 2. Typical garden fruits composite.[a]

Fruit	Form	14-Day quantity, g
Green peppers	Raw	38
Tomatoes	Fresh	296
Tomatoes	Canned	170
Cucumbers	Raw	114
Catsup		128
Pickles		94
Vegetable soup	Canned	163
Tomato soup	Canned	32

[a]Based on FY 1973 Total Diet Survey data of the Food and Drug Administration.

Qualitative evaluation of the data reveals that the pattern of distribution in foods differs substantially among the various metals. Cadmium has the most widespread distribution (Table 3). Specific sources of high concentrations of cadmium are shellfish and some vegetables such as spinach; however, when both food consumption and concentration of the metal are considered, a number of composites provide substantial portions of the cadmium intake. Among these food classes, cereals and grains provided the greatest percentage of total intake.

Table 3. Dietary sources of lead, cadmium, and zinc.

Metal	Major dietary source by food class	% of total dietary intake[a]
Cadmium	Generalized distribution Highest in grain and cereal products	23
Lead	Generalized distribution highest in:	
	Vegetables, all types combined	44
	Fruits and garden fruits	34
Zinc	Dairy products	21
	Meats, fish, and poultry	37
	Grains and cereal products	19

[a]Based on FY 1973 Total Diet Survey data of the Food and Drug Administration.

Lead is also found in a large number of foods. However, fruits and vegetables are the most important sources. While they constitute only 23% of the total diet by weight, they account for 78% of the total lead intake (Table 3). Zinc was present in all the food classes examined. However, dairy products, meat–fish–poultry, and grains and cereals provide 77% of the zinc (Table 3), even though they account for only about half the diet by weight.

Arsenic, which is reported as As_2O_3, has a narrower distribution. In recent years, arsenic occurred most frequently in the meat–fish–poultry composites. Shellfish have been reported to contain generally higher levels, but they are not significant components of the average diet. Patterns of food consumption are such that meat–fish–poultry, dairy products, and grains and cereals are important dietary sources. Of the total arsenic in the diets studied in this program, 92% came from these three food classes (Table 4). With regard to selenium, essentially all selenium in the diet is attributable to the meat–fish–poultry group and the grain-cereal group. These two food classes provide over 99% of the selenium intake (Table 4).

Mercury has the most limited distribution of the metals studied. Virtually all dietary intake of mercury was found in the meat–fish–poultry class and in fish within that class where followup studies were performed (Table 4). This observation is further supported by Simpson et al. (*2*) in a report

Table 4. Dietary sources of arsenic, selenium, and mercury.

Metal	Major dietary source by food class	% of total dietary intake[a]
Arsenic (as As_2O_3)	Dairy products	31
	Meat, fish, and poultry	56
	Grains and cereal products	14
Selenium	Meat, fish, and poultry	38
	Grains and cereal products	62
Mercury (total)	Meat, fish, and poultry	100

[a]Based on FY 1973 Total Diet Survey data of the Food and Drug Administration.

of a study on fish, other staple foods, and total diet composites. The report indicated that, except for fish, mercury levels in foods are generally so low as to be undetected by the usual flameless atomic absorption techniques; neutron activation analysis was necessary to detect any mercury present. The Market Basket surveys as currently performed give a good indication of general trends of distribution of the metals in foods. However, in some cases there may be greater intakes from some food classes than currently apparent because of the detection limits of the chemical methods employed. This problem becomes particularly apparent in the food classes that constitute a large percent of the total diet but may contain low concentrations of some of the metals: dairy products, meat–fish–poultry and beverages.

Detailed data on the findings of heavy metals in various food classes are presented in Tables 5 and 6. In evaluating these data it has been recognized that the use of food composites creates difficulties in determining actual total quantities of metals present in foods. For example, the concentration of lead in many of the composites is very close to the detection limit of the method. Thus, analyses of many of the composites will yield the analytical report of trace or zero. How these traces or zeros are handled in calculations will substantially affect the magnitude of the value reported for total intake. This problem has been discussed in depth in a previous publication (3). A wide range of total lead intakes can be calculated based on various analytical assumptions (Table 7). Among the metals included in the Total Diet Study, this problem is most severe for lead. Cadmium, arsenic, and selenium present far fewer difficulties in this regard because of lower levels of detectability in relation to the levels that occur. The chemical methodology for zinc and mercury is sufficiently reliable at the levels determined that the estimated intakes for these metals are considered to be very accurate approximations of the actual values. Table 8 presents the actual values found in the 1973 Total Diet Survey and the effects of various assumptions on them. Although total exposure to lead may be underestimated, this is not as significant a problem for most of the metals determined in this study.

Table 9 shows yearly values in the metal content of foods, as obtained from the Total Diet Surveys. Although the total dietary lead content is undoubtedly underestimated, the chemical methodology and handling of traces and zeros in calculations has remained constant, and the same is true for other metals. Thus comparisons of yearly changes are meaningful. These data, shown in Ta-

Table 5. Heavy metals findings by food class.

		Heavy metal, % (ppm)[a]					
	Food group	Lead	Cadmium	Zinc	Arsenic (as As_2O_3)	Selenium	Mercury
I	Dairy products	33 (0)	17 (0.005)	100 (4.99)	3 (0.0033)	43 (0)	0 (0)
II	Meats, fish, and poultry	77 (0.013)	40 (0.0093)	100 (24.5)	27 (0.02)	100 (0.21)	97 (0.011)
III	Grains and cereal products	73 (0.10)	97 (0.028)	100 (8.07)	7 (0.003)	100 (0.22)	3 (0)
IV	Potatoes	57 (0.0033)	100 (0.046)	100 (5.9)	3 (0.0033)	23 (0.0033)	0 (0)
V	Leafy vegetables	83 (0.050)	100 (0.051)	100 (2.2)	7 (0)	33 (0)	0 (0)
VI	Legume vegetables	100 (0.26)	33 (0.006)	100 (7.6)	7 (0)	40 (0)	0 (0)
VII	Root vegetables	83 (0.11)	80 (0.021)	100 (2.3)	7 (0)	20 (0.0067)	3 (0)
VIII	Garden fruits	87 (0.12)	80 (0.019)	97 (3.0)	0 (0)	27 (0)	0 (0)
IX	Fruits	73 (0.043)	17 (0.042)	97 (0.88)	3 (0)	13 (0)	0 (0)
X	Oils, fats, and shortenings	50 (0.013)	97 (0.027)	100 (6.23)	7 (0.0033)	43 (0)	3 (0)
XI	Sugars and adjuncts	63 (0.0067)	43 (0.0083)	100 (3.13)	3 (0)	10 (0)	0 (0)
XII	Beverages	17 (0.0033)	17 (0.0057)	97 (0.94)	0 (0)	13 (0)	3 (0)

[a]Listings are presented as % occurrence in the respective food class followed by average ppm level. Traces count as positive occurrence but with no ppm value.

Table 6. Estimated dietary intakes of heavy metals by food class.[a]

		Lead		Cadmium		Zinc		Mercury		Arsenic		Selenium	
		μg/day	% of total	μg/day	% of total	μg/day	% of total	μg/day	% of total	μg/day	% of total	μg/day	% of total
I	Dairy products	0.0	0.0	3.94	7.7	3837	21.4	0.0	0.0	2.34	23.1	0.0	0.0
II	Meat, fish, and poultry	4.00	6.6	2.49	4.9	6660	37.2	2.89	100.0	5.64	55.6	56.3	37.6
III	Grain and cereal	4.16	6.9	11.66	22.8	3370	18.8	0.0	0.0	1.35	13.7	92.5	61.8
IV	Potatoes	0.70	1.2	9.11	17.8	1198	6.7	0.0	0.0	0.64	6.3	0.65	0.4
V	Leafy vegetables	3.03	5.0	3.18	6.2	136	0.8	0.0	0.0	0.0	0.0	0.0	0.0
VI	Legume vegetables	18.80	31.1	0.42	0.8	542	3.0	0.0	0.0	0.0	0.0	0.0	0.0
VII	Root vegetables	3.83	6.4	0.76	1.5	80	0.5	0.0	0.0	0.0	0.0	0.25	0.2
VIII	Garden fruits	11.36	18.8	1.71	3.4	267	1.5	0.0	0.0	0.0	0.0	0.0	0.0
IX	Fruits	9.49	15.7	9.38	18.3	194	1.1	0.0	0.0	0.0	0.0	0.0	0.0
X	Oil and fats	0.67	1.1	1.36	2.7	314	1.8	0.0	0.0	0.17	1.7	0.0	0.0
XI	Sugars and adjuncts	0.55	0.9	0.68	1.3	254	1.4	0.0	0.0	0.0	0.0	0.0	0.0
XII	Beverages	3.81	6.3	6.49	12.7	1066	6.0	0.0	0.0	0.0	0.0	0.0	0.0
	Totals	60.4		51.2		17918 (17.9 mg)		2.89		10.1		149.7	

[a]Based on FY 1973 Total Diet Survey data of the Food and Drug Administration.

Table 7. Effect of various analytical assumptions on calculation of dietary lead content.[a]

Dietary group	Analytical assumption	Total dietary lead intake, μg/day
15–20 year-old males	Zeros and trace amounts not used in calculations	57
15–20 year-old males	Assumed trace = 0.09 ppm	159
15–20 year-old males	Assumed trace = 0.09 ppm Assumed zero = 0.05 ppm	233

[a]From Kolbye et al. (3)

Table 8. Estimates of total daily intake for several trace metals.

Metal	Total daily intake, μg	Minimum quantitation level, ppm	Probable effect of compositing
Lead	60	0.1	Substantial underestimation
Cadmium	51	0.02	Very slight underestimation
Zinc		0.5	Good estimate
Mercury (as total Hg)	2.89	0.01–0.02	Good estimate
Arsenic (as As_2O_3)	10.1	0.1	Minimal effect
Selenium	150	0.1	Minimal effect

ble 9, indicate no trends of increasing or decreasing metal exposure in the diet.

In evaluating the toxicological significance of the various metals, the estimated intake from the Total Diet Study has been compared with the WHO/FAO provisional recommendations of tolerable intakes (*4*) for adults (Table 10). Concerning lead, two points are apparent. Because of underestimation of actual lead intake the per cent of the recommended level is far higher than the 14% which is calculated. However, on the basis of data presented in an earlier paper from our group it is known that the average lead intake is well below

Table 9. Heavy metal content of foods by year.

Metal	Daily intake[a]						
	1968	1969	1970	1971	1972	1973	1974[b]
Cadmium	26	61	38	29	37	51	34
As_2O_3	c	c	c	c	c	10	21
Mercury	ND[d]	ND	ND	ND	3.85	2.89	2.84
Lead	ND	ND	ND	ND	ND	60	90
Selenium	ND	ND	ND	ND	ND	150	169
Zinc	ND	ND	ND	ND	ND	18.0	18.6

[a]Daily intake in μg/day for all except zinc, which is in milligrams per day.
[b]Estimated level.
[c]1965–1970 average = 63 μg/day; 1971–1972 period not calculated at this writing.
[d]Not determined.

the WHO/FAO tolerable level for adults. No WHO/FAO recommendations yet exist for children. For cadmium and mercury the percent of tolerable intake is considered a good estimate. Cadmium is close enough to the tolerable intake so that further increases in the cadmium content of foods should be avoided. Thus far WHO/FAO tolerable intakes for arsenic and selenium have not been established.

Selenium and zinc are both essential nutrients. The National Research Council of the USA recommends an intake of 15 mg zinc/day for adults (*5*). The teenage boy who consumes a large amount of food can easily meet this requirement. However, the recommended dietary allowances for pregnant and lactating women are 20 and 25 mg zinc per day, respectively. With lower calorie intakes and a higher requirement, foods must be carefully selected in order to avoid the problem of an undesirably low zinc intake. National Research Council recommendations on selenium intake have not been set (*5*).

In summary, for the metals included in the Total Diet Study it is generally considered that while the food supply contains less than tolerable intakes of those toxic heavy metals for which recommendations exist, any increases in these trace metal concentrations of food are undesirable.

Table 10. Dietary intakes of heavy metals.

	WHO/FAO provisional tolerated weekly intakes for man[a]		Transformation of WHO/FAO limits to daily basis		FY 1973 Total diet findings		% of tolerable daily intake
	mg/person/week	mg/kg body wt/week	μg/person day	μg/kg body wt./day	μg/person/day	μg/kg body wt./day	
Mercury							
Total mercury	0.3	0.005	42.9	0.71	2.89[b]	0.041	6.7
Methylmercury expressed as mercury	0.2	0.0033	28.6	0.48	—	—	—
Lead	3.0[c]	0.05	429.0	7.15	60.4	0.87	14.1
Cadmium	0.4-0.5	0.0067-0.0083	57.1-71.4	0.95-1.19	51.2	0.73	89.7-71.7
Selenium	Not established		—	—	149.7	2.15	—
Arsenic	Not established		—	—	10.1 (as As_2O_3)	0.15 (as As_2O_3)	—

[a]From 16th Report of Joint FAO/WHO Expert Committee on Food Additives (*4*).
[b]FDA determines total mercury in the total diet study.
[c]WHO/FAO intakes of lead were specifically stated as not being applicable to infants and children.

REFERENCES

1. Food and Drug Administration Compliance Program Guidance Manual, 7320.08. Total Diet Studies—Adult, Washington, D.C., 1974.
2. Simpson, R.E., Horwitz, W., and Roy, C.A. Surveys of mercury levels in fish and other foods. Pestic. Monit. J. 7: 127 (1974).
3. Kolbye, A. C., Mahaffey, K. R., Fiorino, J. A., Corneliussen, P. E., and Jelinek, C. F. Food exposure to lead. Environ. Health Perspect. 7: 65 (1974).
4. 16th Report on Joint FAO/WHO Expert Committee on Food Additives, Geneva, 1972.
5. National Academy of Sciences-National Research Council. Recommended Dietary Allowances. 8th ed., Washington, D.C., 1974.